Journal of Engineering Physics and Thermophysics, Vol. 86, No. 5, September, 2013

KINETIC THEORY OF TRANSFER PROCESSES

MULTICOMPONENT SEPARATION POTENTIAL. ELEMENTARY KINETIC THEORY

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UDC 621.039.31

A molecular model of a multicomponent separation process has been constructed. A general expression for the multicomponent separation potential (value function) has been derived from first principles. The general expression is not tied to a concrete cascade scheme and coincides with the classical expression for the case of a binary mixture. A peculiarity of the separation process, which leads to an infinite value of the potential for pure components of the mixture, has been revealed.

Keywords: *multicomponent separation potential, kinetic theory, isotope separation.*

Studying the multicomponent separation potential is a topical problem that is expediently solved from first principles, i.e., on the basis of the molecular-kinetic model of a separation process without ties to a concrete cascade. This seems correct, since the theory of multicomponent separating cascades is quite complicated and is in the process of development.

The cost of the energy expended on separating a mixture of isotopes, i.e., on changing the concentration of the required isotope in the mixture, is the main item of expenditure of the separation production. The minimum energy expended in the "ideal" process of separation of isotopes is called "separative work." This notion is of special importance in separation theory, since it enables one to compare various methods of isotope separation and to evaluate the expenditure on obtaining a certain concentration of the isotope from the raw material with a prescribed concentration of the components. The separative work ΔU depends just on the initial and final state of the mixture and, for a mixture of two components, is defined as

$$\Delta U = M_{\mathrm{p}} V\left(C_{\mathrm{p}}\right) + M_{\mathrm{w}} V\left(C_{\mathrm{w}}\right) - \left(M_{\mathrm{p}} + M_{\mathrm{w}}\right) V\left(C_0\right)$$

or

$$\Delta U = \Delta U_{\mathrm{p}} + \Delta U_{\mathrm{w}} = M_{\mathrm{p}}\left(V\left(C_{\mathrm{p}}\right) - V\left(C_0\right)\right) + M_{\mathrm{w}}\left(V\left(C_{\mathrm{w}}\right) - V\left(C_0\right)\right).$$

This work is directly proportional to the product of the number of moles M_j of the separated mixture and to the change in a certain function $V(C_j)$ called the separation potential [1] and dependent on the concentration of the mixture C_j. By analogy with the thermodynamic potential, the separation potential $V(C)$ is a dimensionless quantity reflecting the minimum energy that is necessary for changing the concentration of one mole of the mixture regardless of a concrete separation process [2, 3]. The separation potential is also called the "value function," since the product $MV(C)$ reflects the "cost" of the number of moles M of the mixture with concentration C.

The issue of the physical nature of the separation potential and of the method of derivation of an expression for it is still controversial. The classical expression for the separation potential [4], derived by Dirac and Peierls for a binary mixture of isotopes (uranium-235 and uranium-238) on the basis of Dirac′s axioms in the approximation of a small change in the concentration of the mixture, is of the form

$$V\left(C\right) = \left(2C - 1\right) \ln\left(\frac{C}{1 - C}\right). \tag{1}$$

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Translated from Inzhenerno-Fizicheskii Zhurnal, Vol. 86, No. 5, pp. 1140–1147, September–October, 2013. Original article submitted January 14, 2013.

However, the method of derivation of (1) leaves open the issue as to the physical principles of the minimum thermodynamic separative work and to the range of applicability of the separation potential (1) to various separation processes. Furthermore, this expression cannot be directly applied to multicomponent mixtures.

In deriving (1), it was assumed that: 1) V depends just on the mixture′s concentration; 2) the separative work ΔU does not depend on this concentration [5]. The obviousness of these axioms, known as Dirac′s axioms, is different as far as their physical meaning is concerned. Whereas the first axiom is beyond question, since it is only the concentration of the end product that is of interest in the process of separation of the mixture and the expenditure on the separation process must reflect the change in this precisely concentration under the conditions of constancy of the remaining thermodynamic parameters, the second axiom is not as indisputable as this. Generalizations of formula (1) to multicomponent mixtures were repeatedly proposed [6–9], but such generalization on the basis of direct calculation of the elementary molecular-kinetic separation model was not attempted.

Let us consider a ternary mixture of isotopes. The separation potential V allows the computation of the minimum energy expenditure ΔU on separating the original mixture of three components with concentrations C_{01}, C_{02}, and C_{03} into three parts with concentrations C_{11}, C_{12}, C_{13}, C_{21}, C_{22}, C_{23} and C_{31}, C_{32}, C_{33}:

$$\delta U = G_1 V\left(C_{11}, C_{12}, C_{13}\right) + G_2 V\left(C_{21}, C_{22}, C_{23}\right) + G_3 V\left(C_{31}, C_{32}, C_{33}\right) - G_0 V\left(C_{01}, C_{02}, C_{03}\right). \quad (2)$$

Here G_i are the number flow rates (fluxes), where $i = 0$ at entry and $i = 1$, 2, and 3 at exits from the separation plant. This formula is a generalization of the classical expression for the separative work for the case of a ternary mixture. A significant point in formula (2) is the presence of three selection fluxes, G_1, G_2, and G_3, since it is only in the presence of these three fluxes that the ternary mixture can be separated into the components. One mistake of the previous approaches to the problem in question was neglect of this circumstance and deriving the separation potential with just two output fluxes or binary cascades. Although the actual processes of multicomponent separation are frequently effected on binary cascades, it is clear that a binary cascade with intermediate selection represents a separation element with three or more selections. As the number of the components of the mixture increases, the number of selection fluxes in formula (2) must be increased and be equal to the number of the components in the mixture. Aiming at computing the minimum work required for separation of the isotope mixture, we must proceed from an ideal scheme of a separation element, not from the scheme of a concrete cascade. However, this does not mean that the expressions presented below are inapplicable to the separation of multicomponent mixtures in classical binary cascades.

The classical thermodynamic approach to deriving the separation potential is usually reduced to consideration of the entropy of mixing and dates back to Peierls′ classical works [2]. It is stated in this approach that the minimum thermodynamic separative work can be evaluated as the energy of isochoric-isothermal mixing and separation:

$$dE = TdS,$$

where dS is the change in the mixture′s entropy in mixing and separation.

To evaluate the expenditure of energy on the thermodynamic process from the expenditure and release of energy for the opposite thermodynamic process (e.g., to evaluate the energy necessary for separating a mixture from the energy of its mixing), we must have evidence of the thermodynamic reversibility of the process. The sign of reversibility of a certain process is formulated thermodynamically as follows: the entropy change of the process is equal to the quantity of heat δQ supplied to the system, which is divided by the absolute temperature T:

$$dS = \delta Q/T.$$

A sign of an irreversible process is the condition

$$dS > \delta Q/T.$$

Considering the mixture as an isolated system without heat supply ($\delta Q = 0$), we have $dS > \delta Q/T$, i.e., it is impossible, in principle, to effect the process in the opposite direction through the same states of the system and, above all, with the same expenditure of energy that has been released or absorbed in the direct process. In this case it makes no sense to argue about the entropy of mixing $dE = TdS$ as the lower energy limit necessary for separating.

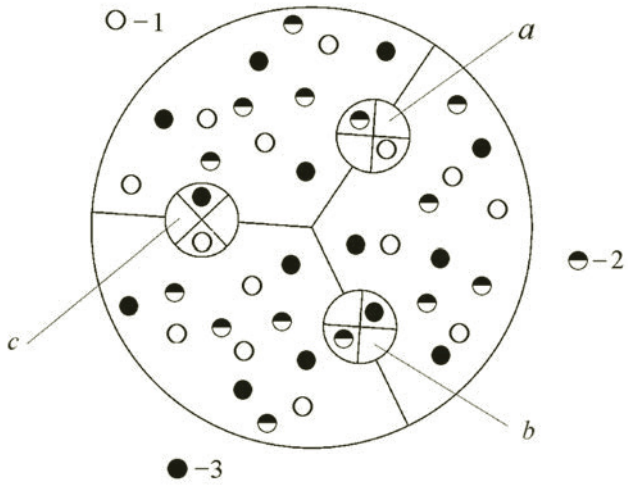

Fig. 1. Molecular model of a three-component separation process: a) exchange device 1 ↔ 2, b) 2 ↔ 3, and c) 1 ↔ 3.

We have proposed, in [10], a simple molecular-kinetic model of a binary separation element, which yields expression (1) for the separation potential. Analogously to the case of a binary mixture, we can consider a molecular model of separation of a multicomponent mixture. The separation of a ternary mixture may be represented by the molecular separation model shown in Fig. 1. The original mixture is divided by impermeable partitions into three volumes. In each partition, there is a certain exchange device effecting selective exchange of molecular pairs between the volumes. The volumes may the unequal. The volume for the enrichment with a corresponding component is proportional to its concentration in the original mixture. Presumably, the volumes remain constant in the process of separation and the total number of particles in each volume is invariable, too, which guarantees the absence of expenditure to do work against pressure forces.

Exchange devices (Fig. 1) ensure paired exchange of molecules of required sorts between two volumes of the mixture. Exchange device 1 ↔ 2 looks for a black and white molecule in volume 1, and it looks for a white molecule in volume 2 and exchanges the pair of molecules between the volumes. The other two exchange devices operate analogously.

We assume that the exchange is effected so that only the required molecule enters the desired volume. Clearly, in operation of such exchange devices, the mixture can be enriched with the desired molecules in each of the three volumes and each action of the exchange device leads to a separation event. A peculiarity of this scheme is the contact of each volume with each volume. Clearly enough, this is also possible for a quaternary mixture. For a larger number of the components, displaying this scheme involves topological difficulties but such paired connections of each volume to each volume may also be presented for an arbitrary number of components.

Analyzing the schematic presented in Fig. 1, conclude that the power is independent of the operation of exchange devices. Here it should be understood that the expenditure on exchanging a molecular pair is determined by the concentration of molecules in the volume from which the molecule is removed [10]. For example, "white" molecules are accumulated in volume 1. But the alteration of the number of "white" molecules by exchange device 1 ↔ 3 will by no means have an effect on the operation of exchange device 1 ↔ 2, since the number and concentration of "black and white" molecules removed by this device from volume 1 do not change because of this. Analogously, the concentration of "white" molecules in volume 2, too, is independent of the operation of exchange device 2 ↔ 3.

The concentration of "black and white" molecules in volume 1 and of "white" molecules in volume 2 determine the expenditure of energy of transfer in exchange device 1 ↔ 2. The aforesaid means that the energy necessary for searching for the "black and white" molecules in volume 1 remains constant. Similarly the energy necessary for searching for the "white" molecules in volume 2 remains constant, i.e., the operation of exchange device 1 ↔ 2 is independent of whether the other two exchangers operate.

Actually, this scheme ensures the operation of a ternary separator as three binary separators operating independently. Since this scheme can separate a mixture into pure components and does not effect "useless" particle exchanges, it may be stated that it consumes the smallest possible quantity of energy. Consequently, the energy expended by an exchange device in

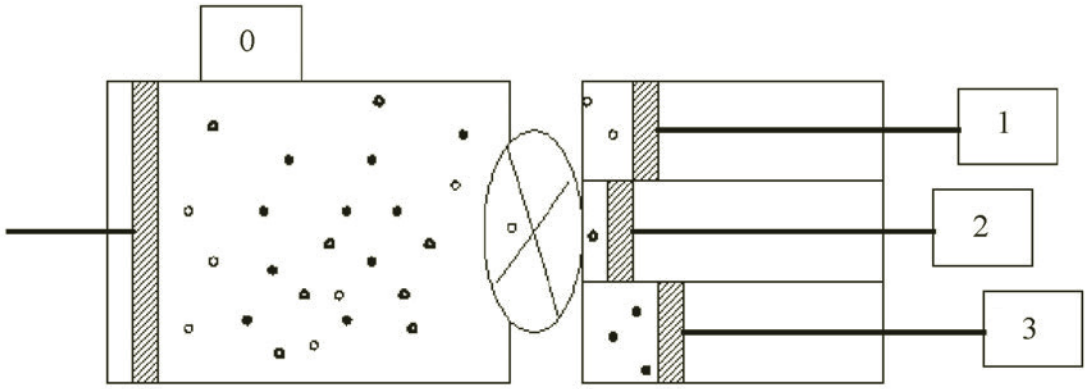

Fig. 2. Entropy separator: 0) volume of the original mixture; 1–3) resulting volumes.

the model in question is the minimum necessary for separating a ternary mixture, and it may be qualified as the potential of the separation process. As has been shown in [10], separation devices of the type of a gas centrifuge and a diffusion separation filter correspond to the model in Fig. 1.

A schematic of an entropy separator [10] whose energy expenditure corresponds to the mixing entropy is shown in Fig. 2. A selector removes a random molecule from the original mixture (volume 0) and transports it in accordance with its sort into one of the three resulting volumes (1, 2, or 3). Pistons move so as to ensure the constancy of pressure in the volumes. Conceivably, the model of such separation might be realized with laser technologies; however, none of the technologies based on molecular diffusion can ensure a finite expenditure of energy in separating totally.

The energy required for the operation of the model in Fig. 1 can be represented as the sum of three independent terms, i.e., of the energies required for the operation of three binary separators. Since the exchange devices operate independently, the process of separation can be represented as the successive action of these devices, i.e., first, the operation of exchange device 1 ↔ 2 is fully completed, then the operation of exchange device 2 ↔ 3, and finally that of exchange device 1 ↔ 3. We emphasize that the operation of exchange devices in the scheme in question is absolutely independent and it makes no difference whether they operate successively or simultaneously. Since the devices are identical, we need only compute the expenditure of energy for one of them, e.g., for exchange device 1 ↔ 2.

As has been indicated in [10], the expenditure of energy on exchanging a pair of particles consists of two parts: 1) the expenditure on searching for the required pair of particles ε_f and 2) the expenditure on exchanging particles ε_t. Since the mixture is assumed to be thermodynamically equilibrium, the probability that a particle of the i sort will be selected in the volume is N_i/N.

An average of [10]

$$n_i = \frac{N - N_i}{N_i} + 1 = \frac{1}{C_i}$$

samples from the volume concentration C_i should be made to find a molecule of the i sort in it. This is independent of the relation of the concentrations of the other two components and hence of the operation of the remaining exchangers. The expenditure of energy on changing the concentration C_i in the volume by a finite value will be [10]

$$\Delta E_{\text{f}i} = \left| \int_{C_{i,0}}^{C_i} \frac{kT}{C}\, dC \right| = kT \left| \ln\left(\frac{C_i}{C_{0,i}}\right) \right|.$$

Thus, the total expenditure on searching for a molecular pair by exchange device 1↔2 will be

$$\Delta E_{\text{f}1} + \Delta E_{\text{f}2} = kT \left(\left| \ln\left(\frac{C_1}{C_{0,1}}\right) \right| + \left| \ln\left(\frac{C_2}{C_{0,2}}\right) \right| \right),$$

the concentration C_1 corresponds to a particle of sort 1 in volume 2, and the concentration C_2 corresponds to a particle of sort 2 in volume 1, but at the instant of the beginning of separation, the concentrations in all volumes are equal. Also, this can be written as

$$\Delta E_{f12} = E_{f12}\left(C_1, C_2\right) - E_{f12}\left(C_{0,1}, C_{0,2}\right),$$

where

$$E_{f12}\left(C_1, C_2\right) = -kT\left(\ln\left(C_1\right) + \ln\left(C_2\right)\right).$$

The exchange of the selected pair of particles $1 \leftrightarrow 2$ consists of: a) removing one selected particle of sort 2 from volume 1 and placing a particle removed from volume 2 to volume 1; b) removing one selected particle of sort 1 from volume 2 and placing the particle removed from volume 1 in volume 2.

Let us consider the expenditure of energy ε_1 on removing one particle of sort 2 from volume 1. Within the model of a mixture of perfect gases, this energy is independent of the sort of particle and is a constant, i.e., for any particle, we can write $\varepsilon_1 = A$, where A is the constant. Then the expenditure on removing an infinitely small number of particles dn of sort 2 from volume 1 will be equal to $d\varepsilon_1 = Adn$. We express the energy ε_1 as a function of the concentration C of particles of sort 2. The concentration change dC is related to the decrease, by dn, in the number of particles n of sort 2 in volume 1 with N particles by

$$dC = C - C' = \frac{n}{N} - \frac{n - dn}{N - dn}.$$

Hence we can express dn by dC:

$$dn = \frac{N^2 dC}{N - n + NdC} = \frac{NdC}{1 - C + dC}.$$

Neglecting higher degrees of dC, we obtain

$$dn = N\frac{dC}{1 - C},$$

which yields that

$$d\varepsilon_1 = AN\frac{dC}{1 - C}.$$

Integration of this expression will give

$$\varepsilon_1\left(C\right) = -AN\ln\left(1 - C\right).$$

Let us consider the expenditure of energy ε_2 on placing the particle removed from volume 2 in volume 1. Within the framework of the model of a mixture of perfect gases, this energy is independent of the sort of particle and is a constant, i.e., we can write $\varepsilon_2 = B$ for any particle, where B is the constant. Then the expenditure on placing an infinitely small number of particles dn of sort 1 in volume 1 will be equal to $d\varepsilon_2 = Bdn$. Assuming that the number dn of placed particles is exactly equal to the number of removed particles, we express the sought energy as the function of the concentration C of particles of sort 2 in volume 1. The concentration change dC is related to dn by

$$d\left(1 - C\right) = \frac{N - n}{N} - \frac{N - n + dn}{N + dn}.$$

Hence we can express dn by dC:

$$dn = \frac{N^2 dC}{n - NdC} = \frac{NdC}{C - dC}.$$

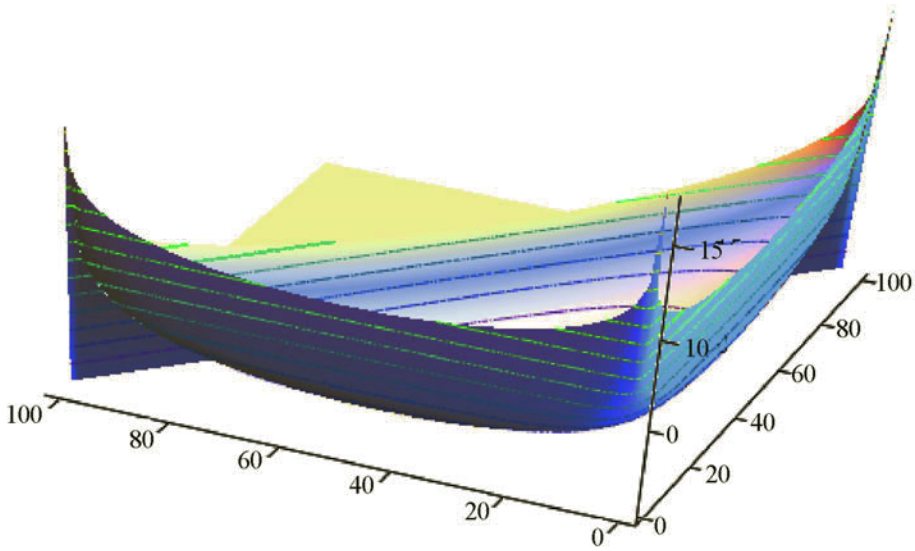

Fig. 3. Schematic representation of the three-component separation potential (4).

Neglecting higher degrees of dC, we obtain

$$dn = N\frac{dC}{C},$$

which yields that

$$d\varepsilon_2 = BN\frac{dC}{C} \quad \text{and} \quad \varepsilon_2(C) = BN\ln(C).$$

We determine what the energies ε_1 and ε_2 are expended on. Since the process of removal and insertion of molecules is assumed to be isothermal, PV = const, clearly, it is only the pressure P that changes in the volume V, i.e., the energy is expended on doing work against pressure forces. In this case the constants A and B in the expressions for ε_1 and ε_2 are equal and we can write

$$\varepsilon_t(C) = \varepsilon_1 + \varepsilon_2 = AN[\ln(C) - \ln(1 - C)].$$

The energy $\varepsilon_t(C)$ is expended on changing one pair of particles, i.e., on an infinitely small change in the concentration C. To pass to determining the energy expended on changing finitely the concentration C, we must integrate the expression for $\varepsilon_t(C)$:

$$E_t(C) = AN\int \ln(C) - \ln(1 - C)\,dC = AN\left[C\ln(C) + (1 - C)\ln(1 - C)\right].$$

It turns out that the expenditure of energy is expressed by a formula coincident with the expression for the entropy of a binary mixture. This was to be expected, since a particle of a specific sort is removed from the volume, whereas all the remaining particles (of the other two sorts) are indistinguishable, i.e., the mixture is binary, in fact. By virtue of the expressed analogy between the last expression and the entropy of a binary mixture, it may be assumed that the amplitude factor is $AN = kT$. Finally, we obtain

$$E_t(C) = kT[C\ln(C) + (1 - C)\ln(1 - C)]$$

or, for the complete cycle of exchange of a pair of particles:

$$E_t(C_1, C_2) = kT\left[C_1\ln(C_1) + (1 - C_1)\ln(1 - C_1) + C_2\ln(C_2) + (1 - C_2)\ln(1 - C_2)\right].$$

At the instant of the beginning of separation, the concentrations in all volumes are equal for all the three exchange devices. Consequently, we can write the following expressions for exchange devices

1↔2:

$$E\left(C_1, C_2\right) = E_{\mathrm{f}}\left(C_1, C_2\right) + E_{\mathrm{t}}\left(C_1, C_2\right) = kT\left(\left(C_1 - 1\right)\ln\left(\frac{C_1}{1 - C_1}\right) + \left(C_2 - 1\right)\ln\left(\frac{C_2}{1 - C_2}\right)\right),$$

1↔3:

$$E\left(C_1, C_3\right) = kT\left(\left(C_1 - 1\right)\ln\left(\frac{C_1}{1 - C_1}\right) + \left(C_3 - 1\right)\ln\left(\frac{C_3}{1 - C_3}\right)\right),$$

and 2↔3:

$$E\left(C_2, C_3\right) = kT\left(\left(C_2 - 1\right)\ln\left(\frac{C_2}{1 - C_2}\right) + \left(C_3 - 1\right)\ln\left(\frac{C_3}{1 - C_3}\right)\right).$$

The total expenditure of energy on separating is equal to

$$\begin{aligned} E\left(C_1, C_2, C_3\right) &= E\left(C_1, C_2\right) + E\left(C_1, C_3\right) + E\left(C_2, C_3\right) \\ &= 2kT\left(\left(C_1 - 1\right)\ln\left(\frac{C_1}{1 - C_1}\right) + \left(C_2 - 1\right)\ln\left(\frac{C_2}{1 - C_2}\right) + \left(C_3 - 1\right)\ln\left(\frac{C_3}{1 - C_3}\right)\right). \end{aligned} \tag{3}$$

It is precisely the function $E(C_1, C_2, C_3)$ that is the separation potential of the ternary mixture. In the case of the absence of the third component, $E(C_1, C_2\,(C_3 = 0))$, the function of (3) is of the form

$$E\left(C_1, C_2\right) = kT\left(\left(C_1 - 1\right)\ln\left(\frac{C_1}{1 - C_1}\right) + \left(C_2 - 1\right)\ln\left(\frac{C_2}{1 - C_2}\right)\right).$$

This expression with account of $C = 1 - C_2$ is identical to the classical value function of a binary mixture (1). The potential (3) in dimensionless form and with the point $C_1 = C_2 = C_3 = 1/3$ reduced to zero can be written as

$$V\left(C_1, C_2, C_3\right) = \left(C_1 - 1\right)\ln\left(\frac{C_1}{1 - C_1}\right) + \left(C_2 - 1\right)\ln\left(\frac{C_2}{1 - C_2}\right) + \left(C_3 - 1\right)\ln\left(\frac{C_3}{1 - C_3}\right) - \ln\,(4). \tag{4}$$

The function of (4) is shown in Fig. 3. Similarly to the binary case, expression (4) has its minimum at the point of equality of the concentrations of the components $C_1 = C_2 = C_3 = 1/3$ and infinitely grows when any one of the concentrations tends to zero.

Expression (3) is easily generalized to the case of an arbitrary number of the mixture′s components:

$$E\left(C_1, C_2, ..., C_n\right) = \left(n - 1\right)kT\sum_{i=1}^{n}\left(C_i - 1\right)\ln\left(\frac{C_i}{1 - C_i}\right). \tag{5}$$

Since the amplitude factor before the sum in expression (5) is insignificant (of practical interest is only the relative value $E(C_1, C_2, \ldots, C_n)/ E(C_{0,1}, C_{0,2}, \ldots, C_{0,n})$), the multicomponent value function can be written as follows:

$$V\left(C_1, C_2, ..., C_n\right) = \sum_{i=1}^{n}\left(C_i - 1\right)\ln\left(\frac{C_i}{1 - C_i}\right). \tag{6}$$

The expression for the multicomponent value function differs from the variants proposed earlier [6–9]. The value of the mixture, according to (6), is determined by the sum of the terms, each depending just on the concentration of one component, whose number is equal to the number of the components of the mixture. The term of the sum of (6) tends to

infinity if the corresponding concentration tends to zero. In this sense, each term of expression (6) may be considered as a certain minimum thermodynamic work necessary for freeing the mixture from this component.

Expression (6) is a generalization of the classical binary value function (1) which can also be written in the form of (6). The multicomponent value function (6) can also be used for binary separation elements (cascades) separating a multicomponent mixture. In this case only two selection fluxes in formula (2) are nonzero.

The issue of whether the classical formula $\Delta U = G_0\omega(\alpha, \beta, \gamma)$ can be obtained, with the potential (6), for the separative work of a multicomponent element, i.e., whether the separative work can be expressed in a form dependent just on the separation coefficients, is still an open issue. Here $\omega(\alpha, \beta, \gamma)$ is a certain function of the separation coefficients of the element α, β, and γ.

The infinite energy expenditure in separating totally is determined by the assumption that the original mixture is separated into a number of volumes equal to the number of components. Any separation process using several volumes and particle exchange between them leads to repeated involvement of the particles in the separation and to an infinite expenditure of energy in attempting to separate the mixture totally. The reason for this infinity is the reinvolvement of the particles in the process of separation.

This work was carried out with financial support provided by the B. N. Eltsin Ural Federal University to young scientists within the framework of implementation of the Program of Development of the University.

NOTATION

A and *B*, constants; C_i, number concentration of the *i*th component of the mixture, liters/m^3; E_{fi}, expenditure of energy on changing the concentration of the *i*th component in the finite volume of the mixture, J; E_t, expenditure of energy on exchanging particles after the change in the concentration of the *i*th component in the finite volume of the mixture, J;G_i, fluxes of selection of the *i*th component, number flow rate, liter/s; $k = 1.38\cdot10^{-23}$, Boltzmann constant, J/K; *M* and M_i, number of moles of the mixture and of the *i*th component of the mixture, moles; *N* and N_i, number of particles in the mixture and of the *i*th component in the mixture; n_i, average number of samples of *i* particles from the finite volume of the mixture; *Q*, heat supplied to the system J; *S*, entropy of the mixture, J/K; *T*, temperature, K; ΔU, separative power, J; $V(C)$, separation potential (value function); ε_f and ε_t, expenditure of energy on searching for the required pair of particles in the finite volume of the mixture and on exchanging the particle pair in this volume, J. Subscripts: *i*, component number; *j*, flux number; p, selection flux (product); w, waste flux (waste); 0, original mixture or feeding flux of the mixture; f, finite, final; t, transfer, exchange.

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